

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of : Mitsushi ITANO, et al.

Application No.: 10/527,202      Art Unit: 1792

Filed: May 10, 2005      Examiner: Tabassom T. Tadayyon-Eslami

For: ETCHANT AND ETCHING METHOD

Commissioner for Patents  
PO Box 1450  
Alexandria, VA 22313-1450  
Sir:

**DECLARATION UNDER 37 C.F.R. § 1.132**

I, Daisuke WATANABE, do hereby make the following declaration:

1. I am a Japanese citizen, residing at c/o Yodogawa Seisakusho, DAIKIN INDUSTRIES, LTD., 1-1, Nishihitotsuya, Settsu-shi Osaka 566-0044, Japan.
2. I graduated from Kyoto Sangyo University, Faculty of Science, Department of Physics in March 1997. I also graduated from the Graduate School of Osaka University, and received a Master's Degree in Physics in March 2000.
3. I began my employment with DAIKIN INDUSTRIES, LTD, the assignee of the above-identified application, in April 2002. Since 2002, I have been engaged in the research and development of wet chemical for semiconductor device processing.
4. I am one of the named inventors of the above-identified application, and am familiar with the subject

matter of said application as well as the disclosures in the cited references.

5. In order to compare the present invention and the teaching other than the invention, I have conducted the following experiments.

#### Experiment

The object of the experiment was to evaluate the etching effect of the etching solutions recited in the present invention (the following Examples A and B) with the etching solutions not described as the present invention (the following Comparative Examples C, D, E, F, G and H).

##### (1) Example A

The procedure described in Example 7 of the present invention was repeated. The ratio of components in the etching solution of Example A was 20 mass % of HF: 80 mass % of monoglyme: 0 mass % of water.

##### (2) Example B

The procedure described in Example 7 of the present invention was repeated except that the ratio of components in the etching solution was arranged as follows: 20 mass % of HF: 75 mass % of monoglyme: 5 mass % of water.

##### (3) Comparative Example C

The procedure described in Example 7 of the present invention was repeated except that the ratio of components in the etching solution was arranged as follows: 20 mass % of HF: 69 mass % of monoglyme: 11 mass % of water.

##### (4) Comparative Example D

The procedure described in Example 7 of the present invention was repeated except that the ratio of components in the etching

solution was arranged as follows: 20 mass % of HF: 60 mass % of monoglyme: 20 mass % of water.

(5) Comparative Example E

The procedure described in Example 7 of the present invention was repeated except that the ratio of components in the etching solution was arranged as follows: 20 mass % of HF: 50 mass % of monoglyme: 30 mass % of water.

(6) Comparative Example F

The procedure described in Example 7 of the present invention was repeated except that the ratio of components in the etching solution was arranged as follows: 20 mass % of HF: 30 mass % of monoglyme: 50 mass % of water.

(7) Comparative Example G

The procedure described in Example 7 of the present invention was repeated except that the ratio of components in the etching solution was arranged as follows: 20 mass % of HF: 10 mass % of monoglyme: 70 mass % of water.

(8) Comparative Example H

The procedure described in Example 7 of the present invention was repeated except that the ratio of components in the etching solution was arranged as follows: 20 mass % of HF: 0 mass % of monoglyme: 80 mass % of water.

Conditions of the experiment

The etching rates and etching selectivity of each solution were measured for each test substrate comprising, on a silicon substrate, (1) a hafnium oxide silicate film formed by MOCVD, and annealed (MOCVD HfSiO Anneal), and (2) a thermal oxide (THOX) film, respectively.

Etching was conducted by dipping each test substrate of Examples A and B, and Comparative Examples C, D, E, F, G, and H into an

etching solution for one minute. The results are shown below in Table 1.

The conditions of this experiment were the same as that of the Examples described in the present specification except that each test substrate was dipped into an etching solution for one minute.

[Table 1]

	HF (mass%)	Monoglyme (mass%)	Water concent- ration (mass%)	Etching temperature (°C )	Etching rate (Å/minutes)		Etching rate ratio (THOX/HfSiO)
					MOCVD HfSiO Anneal	THOX	
Ex. A	20%	80%	0%	23	9	2	0.22
Ex. B	20%	75%	5%	23	22	23	1.0
Com. Ex. C	20%	69%	11%	23	26	112	4
Com. Ex. D	20%	60%	20%	23	30	277	9
Com. Ex. E	20%	50%	30%	23	32	510	15
Com. Ex. F	20%	30%	50%	23	33	1100	26
Com. Ex. G	20%	10%	70%	23	34	1600	43
Com. Ex. H	20%	0%	80%	23	35	2000	57

#### Consideration on the results of experiment

The etching solutions of Examples A and B according to the present invention contain the compounds as shown in Table 1, in the ratio of HF to the specific ether compound (monoglyme) to water of 3 mass % or greater: 75 to 97 mass %: 5 mass % or less, achieving excellent etching rates. More specifically, the etching rate ratios (THOX/HfSiO) show an unexpectedly excellent value of 1.0 or less.

Accordingly, it is clear that the etching solution of the present invention can etch a High-K film, while greatly suppressing the silicon oxide film etching rate.

6. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of any patent issued on this application.

Date: February 19, 2008 Daisuke Watanabe  
Daisuke WATANABE